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Gas Turbine Engine Non-volatile Particulate Matter mass emissions: Correlation with Smoke number for Conventional and Alternative Fuel Blends

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17

18 ABSTRACT

19 This study evaluates the relationship between the emissions parameters of smoke number
20 (SN) and mass concentration of non-volatile particulate matter (nvPM) in the exhaust of a gas
21 turbine engine for a conventional Jet A-1 and a number of alternative fuel blends. The data
22 demonstrate the significant impact of fuel composition on the emissions, and highlight the
23 magnitude of the fuel-induced uncertainty for both SN within the Emissions Data Bank, as
24 well as nvPM mass within the new regulatory standard under development. Notwithstanding
25 these substantial differences, the data show correlation between SN and nvPM mass
26 concentration still adheres to the First Order Approximation (FOA3), and this agreement is
27 maintained over a wide range of fuel compositions. Hence the data support the supposition
28 that the FOA3 is applicable to engines burning both conventional and alternative fuel blends
29 without adaption or modification. The chemical composition of the fuel is shown to impact
30 mass and number concentration as well as geometric mean diameter of the emitted nvPM,
31 however the data do not support assertions that the emissions of black carbon with small
32 mean diameter will result in significant deviations from FOA3.

33

34 INTRODUCTION

35 Emissions from aircraft gas turbine engines include the combustion products carbon
36 dioxide (CO₂) and water (H₂O), combustion by-products: oxides of nitrogen (NO_x) and
37 products of incomplete combustion: carbon monoxide (CO), unburned hydrocarbons (UHC)
38 and soot aerosol (or black carbon, BC). Each of these species are produced in different
39 relative proportions and all impact or contribute to climate forcing and degradation of air
40 quality^{1,2,3,4}.

41 The International Civil Aviation Organization (ICAO) sets regulatory standards for NO_x,
42 CO, UHC and Smoke Number, which are reported for all certified aircraft engine types
43 >26.7kN thrust in the Emissions DataBank⁵ (EDB). Emissions of BC are not currently
44 reported within the ICAO EDB, but may be inferred through the surrogate measurement of
45 Smoke Number (SN) at specific thrust settings that correspond to those used in the Landing
46 and Take-Off (LTO) cycle. SN is an optically based method that quantifies the change in the
47 reflectance of a Whatman #4 filter paper after sampling a fixed mass of engine exhaust per
48 unit area at a given temperature⁶. The ICAO regulation of SN was originally introduced in
49 1981 as means to quantify aircraft exhaust plume visibility and to act as a driver to reduce
50 emissions. SN does not provide a characterization of BC emissions in terms of mass and
51 number concentration, size distribution, or chemical composition, and given its proxy nature,
52 cannot be used to directly determine the environmental impacts of aviation. Currently, there
53 is an initiative within ICAO to replace the SN with a regulatory measurement methodology
54 for non-volatile particulate matter (nvPM) emissions for aircraft engines certified for use in
55 the commercial sector. In the meantime, SN remains the only measurement whereby BC
56 emissions can be estimated for environmental assessment activities.

57 A number of studies have reported the correlation between SN and mass concentration of
58 black carbon (C(BC)) using a range of different hardware: Champagne⁷ reports a correlation
59 derived from exhaust samples extracted from a combustor rig based on a T56 turboprop
60 engine; Whyte⁸ presented a method to convert between SN and C(BC) from a study of
61 kerosene alternative fuels; and Girling et al.⁹ report a correlation from an experimental study
62 using soot generated by a kerosene fuelled smoke generator amongst others^{10,11}. A critical
63 inter-comparison of these and other data, which agree to within 10%, was presented by
64 Wayson et al.¹². These correlations between SN and C(BC) form the basis of a method
65 endorsed by ICAO's Committee for Aviation Environmental Protection to estimate the mass

66 concentration and/or mass-based emission index of BC emitted from aircraft engines, referred
67 to as the ‘First Order Approximation¹³ version 3’ (FOA3). The FOA3 is intended for use as a
68 standard method to estimate PM mass-based emissions from certified commercial aircraft
69 engines within the vicinity of airports, and as an important assessment tool, there is a
70 commitment to improve FOA3 as new data become available until such time that the
71 methodology is rendered obsolete by a fully validated database of PM emission indices for
72 the commercial fleet.

73 Nevertheless, there has been criticism of FOA3, not least because of the potential for the
74 SN measurement to be dependent upon the capture efficiency of the filter and thus particle
75 size distribution of the emitted BC. This potential for error was first alluded to by Dodds et
76 al.¹⁴, followed by Wayson et al.¹², Sevcenco et al.¹⁵, and most recently discussed by Stettler et
77 al.¹⁶, though any suggestion that the SN underestimates C(BC) would mainly be applicable to
78 more recent engine technology due to the reduced mean diameter of the emitted BC.
79 However, as will be demonstrated and discussed later, the correlation between SN and C(BC)
80 remains a good first order approximation, even when the emitted BC particles have a mean
81 geometric diameter of the order of 20nm.

82 Within the emissions inventory and modeling communities, sources of uncertainty in
83 estimating the mass concentration of BC using SN values may arise when there is the need to
84 interpolate between data points at the four specific LTO thrust settings to determine
85 intermediate values, and also more generally, from the error in the reported SN data itself
86 induced by both measurement uncertainty and the use of non-standardized fuel for
87 certification tests on different engine types. Concerning this latter point, the hydrogen to
88 carbon ratio (H:C) and the aromatic content of the fuel used to produce the SN data for the
89 specific engine type and in the specific emissions certification test are (mostly) recorded
90 within the EDB. The spectrum of reported values in the EDB legacy data of 1.85 – 2.00 for

H:C ratio and 11.9% – 22.5% for aromatic content, covers a range that extends slightly beyond the current recommended specification for fuel to be used in aircraft engine emission testing of 1.85 – 1.99 and 15% – 23%, respectively¹⁷. Nevertheless, even the current ‘tightened’ specification envelope allows for considerable variation in fuel properties, such that the known impacts of fuel composition upon SN are ostensibly not considered. The variation in the fuel properties reported within the ICAO EDB reveal that aromatic content may vary by $\pm 3\%$ at a given H:C ratio, and H:C ratio may vary by ± 0.05 at a given aromatic content. And whilst a decrease in aromatic content is generally associated with an increase in H:C ratio, the correlation between these two parameters is generally poor and insufficient to define the fuel.

Since the introduction of SN, engine technology has made significant progress and certified SN’s at take-off power have decreased from the 25 – 35 range in early data, to values for newer engine technology that typically occupy the 0 – 5 range. However, a SN of zero is clearly a problem for the application of FOA3 in air quality and climate models as it implies that the mass concentration of BC is also zero. For these reasons, ICAO has committed to develop a new direct nvPM standard, but with typical engine lifetimes exceeding 20 years, older legacy engines will continue to contribute to overall emission levels and so both SN and FOA3 may not be fully transitioned for some years to come.

The new ICAO regulatory standard under development for the measurement of aircraft gas turbine engine nvPM number and mass-based emissions uses the standard methodology specified in the Society of Automotive Engineers (SAE) Aerospace Information Report (AIR) 6241¹⁸. The development of this standard methodology for engine nvPM emission measurement was born out of the Aircraft Particle Emissions eXperiment (APEX) campaigns and many other similar studies^{19,20,21,22,23,24,25}. These studies highlighted the complexity of BC emissions measurement, and in particular the difficulty in obtaining repeatable and reliable

measurement data. The data presented here have been obtained using the AIR6241 compliant system North American Mobile Reference System that has been developed and robustly characterized over several years through international collaboration²⁶.

The objective of this work was to compare SN measured using a SAE Aerospace Recommended Practice (ARP) 1179d⁶ compliant system with the nvPM mass concentration measured using the SAE AIR6241 compliant system for a conventional Jet A-1 and a number of alternative fuel blends. In contrast to earlier FOA3 analysis where the correlation was examined in terms of engine technology applicability, here we analyze the FOA3 correlation from a fuel composition perspective. The gas turbine engine used in this study, a Garrett Honeywell GTCP85-129 auxiliary power unit (APU), is not included within the EDB as its rated output is <26.7kN. It is however a suitably close analogue to aircraft main engines that provides a good model soot aerosol, and advances the methodologies previously used in the development of the FOA3.

The chemical composition of the test fuels was managed by introducing various blends of Jet A-1 and a Used Cooking Oil derived Hydrotreated Esters and Fatty Acids (UCO-HEFA) kerosene. A complete range of fuels was investigated to allow full characterization of the data within the FOA3 model, from low blend ratios (0 – 20%) that may be considered to be essentially Jet A-1 variants, through to very high blend ratios that are distinctly paraffinic and alternative in composition. Through this careful management of the fuel composition, it was possible to vary SN in the range from 4 to >40, although the size distribution of the soot aerosol is also present as a co-variable.

The significant impact of fuel aromatic content and/or fuel H:C ratio on nvPM emissions and measured SN, is highly relevant to both the recommended specification for fuel to be used in aircraft engine certification testing and the downstream effect on accurate emission estimates due to regional variability in commercially available aviation fuel. Furthermore, the

potential impact of fuel compositional change becomes considerably more pronounced and pertinent within the context of alternative fuels, and to the projected scale-up of sustainable alternative aviation fuel use (eg. EU Flightpath 2020²⁷), together with future fuel certification, fuel diversification, and long-term fuel security. Sustainable alternative fuels are anticipated to play a sizeable role in decarbonizing the aviation industry, and currently there are no methods to quantify the much-reduced atmospheric burden of BC that results from their use²⁸. Any future update to FOA3 may need to incorporate a SN-fuel composition response function.

BACKGROUND

Soot aerosol

Unfortunately the term ‘soot aerosol’ is rather imprecise in its definition, and terms such as particulate matter, soot, black carbon, graphitic carbon, refractive carbon and non-volatile particulate matter are often used synonymously. On occasions even the term carbon black is used, even though this is distinct in that it is a manufactured product²⁹. Efforts to develop precise nomenclature to distinguish between these terms are on-going, but these are often based on particular measurement techniques or light-absorbing properties^{30,31,32,33} and lack universal acceptance.

In recent years, the term black carbon (BC) has gained widespread usage within the climate and emissions measurement communities, although it is recognized that BC is in itself a generic term that describes a wide range of carbonaceous combustion derived substances from partly charred residues to highly graphitized soot³⁴. BC particles have highly variable physical properties and chemical compositions that very much depend upon their source^{35,36}. Indeed the disparate nature of BC from different sources is well established and has even been used in source apportionment studies. Physical properties such as size, morphology,

heterogeneity, surface area, isotopic ratio and density are all variable, as is chemical composition with solvent extractable organic matter, and total carbon sometimes being primarily elemental carbon (EC), but more often existing as complex mixtures of EC and organic carbon (OC), with volatile and semi-volatile hydrocarbons, and other non-carbon species such as ionic species, sulphates, moisture and trace metals^{37,38,39,40}.

Laboratory-generated ultrafine EC particles such as those created in a diffusion flame are yet another distinct form of carbonaceous material. Overall, scientific studies need to clearly distinguish between these highly disparate EC-containing particles with care and precision to forestall the unwarranted extrapolation of properties and the transposition of inappropriate study conclusions from one material to another. Black carbon from one combustion source is not necessarily a model particle that is representative of the characteristics of an entirely different combustion source.

The focus of this work is to evaluate the correlation between current and forthcoming regulated measurement techniques using the soot aerosol emitted from a gas turbine engine burning a conventional Jet A-1 and a number of alternative fuel blends. The precise bounds and classification of the emitted soot aerosol is therefore operationally defined by the measurement technique employed. Within this text, the term black carbon is used to define the measurand associated with the measurement of smoke number through SAE ARP1179d, whilst the term non-volatile particulate matter is used to define the measurand associated with the measurement of mass concentration, number concentration and size distribution through SAE AIR6241, although it is recognized that size distribution is not a formal part of this standard. The term soot aerosol is used elsewhere in the broader discussion to represent less defined states.

Impact of fuel chemistry on soot aerosol formation

191 Aviation Jet A-1 is a complex cocktail of thousands of different hydrocarbon component
192 molecules, though these molecules are often categorized into four principal groupings: n-
193 paraffins, iso-paraffins, cyclo-paraffins and aromatics⁴¹. The former two groupings of n- and
194 iso-alkanes typically dominate the class composition of all-fit-for-purpose petroleum derived
195 fuels⁴².

196 Variability in the chemical composition of Jet A-1 (and other kerosene specifications such
197 as Jet A, JP4, JP8, etc.) over both region and time is commonplace. The extent of this
198 variation is largely reflective of variability in the feedstock crude and localized demand for
199 other petrochemical distillation fractions. It is assessed on a regional level within fuel survey
200 data such as Rickard⁴³ or the Petroleum Quality Information System⁴⁴ (PQIS). Furthermore,
201 this diversity in the chemical composition of aviation kerosene is set to increase as alternative
202 fuels from a variety of sources enter the market as blend components or substitute fuels.

203 Perhaps the most notable impact of low aromatic kerosene fuels, including Jet A-1 blended
204 with Fischer-Tropsch (F-T) or Hydro-processed Esters and Fatty Acids (HEFA) alternative
205 fuels, is the very strong reduction in black carbon emissions^{45,46,47,48,49,50,51}. For example, the
206 Alternative Aviation Fuel Experiment (AAFEX) study using a CFM56-2C1 engine reported
207 concentrations of BC at the engine exit nozzle may be reduced by as much as 90% using F-T
208 fuels⁵⁰. These reductions affect the mass concentration, number concentration and size of the
209 emitted BC aerosol^{46,47,51}. A detailed evaluation of the impact of small variations in the Jet A-
210 1 / HEFA fuel blend ratio on the emission of nvPM is given in Lobo⁵². Evidence that the
211 reduction in soot aerosol occurs due to the lower aromatic content of the fuel is becoming
212 established, and aromatics are attributed as the class of compounds that primarily influence
213 the tendency to form BC and soot precursors during combustion^{42,45,53}. For example,
214 DeWitt⁴⁵ in an investigation of fuel composition, material compatibility and its relation to
215 emission characteristics showed that BC emissions increase with both increasing fuel

aromatic content and increased aromatic molecular weight when evaluated in a T63 turbo shaft engine. This increase in BC emissions was attributed to an increase in soot precursors.

FOA3: smoke number – mass concentration correlation model

The FOA3 model endorsed by ICAO, is often used to predict the mass concentration of BC in the exhaust emissions of a gas turbine engine from the surrogate smoke number measurement¹³. Such data is routinely required by atmospheric modelers and for the development of emission inventories.

For an engine with $SN < 30$, the mass concentration of BC (mg/m^3) is predicted from the measured smoke number using the following FOA3 equation¹²:

$$C(BC) = 0.0694 (SN)^{1.24} \quad (1)$$

Whereas for $SN > 30$, the mass concentration of BC (mg/m^3) is predicted from the measured smoke number and using the following FOA3 equation¹²:

$$C(BC) = 0.0297 (SN)^2 - 1.802(SN) + 31.94 \quad (2)$$

In both of these equations, $C(BC)$ is reported at standard temperature (273.15 K) and pressure (101.325 kPa), and the bounds of uncertainty for the correlation are dominated by the error in the measurement of the SN as errors in measurement of mass concentration are small in comparison¹².

EXPERIMENTAL METHOD

Gas turbine engine & operating conditions

The Garrett Honeywell GTCP85-129 gas turbine engine used in this study is often operated as an auxiliary power unit (APU) on Boeing 737 aircraft. APU gas turbine engines offer a good model of aircraft main engine combustion characteristics whilst being considerably more manageable and less costly to operate.

241 In this work, three APU operating conditions were investigated: No Load (NL),
242 Environmental Control Systems (ECS), and Main Engine Start (MES). These conditions
243 correspond to the normal operating conditions for an APU. For each experimental run the
244 APU was put through a warm up sequence using Jet A-1 before switching to the test fuel
245 without interruption, and then stabilizing at the first condition. The test matrix followed a
246 successive step down in power from MES to ECS to NL condition, which represented 1 test
247 cycle. For each of the fuel blends evaluated, this test cycle was twice sequenced without
248 shutdown. The sequence stepped down in power to minimize possible differences in
249 operating temperature and therefore potential differences in the fuel vaporization rates that
250 could feasibly manifest themselves as measurement uncertainties. For each engine condition,
251 the emissions data were recorded over a 6 minute window once the APU was determined to
252 be stable (ie. when engine EGT, RPM, and fuel flow were established as consistent).

253 The different fuel blends of Jet A-1 and Used Cooking Oil based HEFA (UCO-HEFA)
254 used for the study were selected at random to mitigate possible systematic bias and drift.
255 Experimental runs with Jet A-1 were conducted at the beginning and end of the study, as well
256 as several times in between runs with different fuel blends to reaffirm baseline conditions.
257 Engine parameters such as fuel flow rate, RPM, air fuel ratio, and exhaust gas temperature
258 were also recorded. The engine was very stable at each operating condition and the
259 reproducibility of engine parameters was good due to the on-board engine management
260 system.

261 Ambient conditions of temperature, pressure, and relative humidity were also recorded
262 throughout, and the range of values for these parameters was: 14.0 – 20.6 °C, 102.47 – 103.11
263 kPa, and 61 – 85%, respectively.

264

265 **Sampling system and instrumentation**

Two identical and almost collocated single-point probes, one for gaseous emissions and SN measurement, and the second for nvPM emissions measurement were placed within $\frac{1}{2}$ nozzle diameter of the engine exit plane (~ 15 cm).

The sample line for gaseous emissions and SN was compliant with the specifications in ICAO¹⁷ Annex 16 Volume 2 and maintained at a temperature of 160°C. Gaseous species were determined using a Binos Non-Dispersive Infrared Sensor (CO), a Signal Flame Ionization Detector (UHC), and an Eco Physics Chemi-Luminescence Analyser (NO_x), each using appropriate span and zero gases between measurements. The SN was determined in accordance with SAE ARP1179d⁶ using a Richard Oliver smoke meter to collect at least three filter samples for each fuel and at each engine condition. The reflectance of the filter samples was determined pre and post sampling using a BOSCH reflectometer. Reported SN data are the arithmetic mean of measurements from 6 filters (2 test cycles x 3 filters at each condition) and uncertainty is conservatively estimated as ± 2 SN. This estimate of uncertainty is consistent with the measured variability, with due recognition that the accuracy of an individual SN measurement is considered to be ± 3 SN⁶.

The nvPM emissions were measured using the AIR6241 compliant North American mobile reference system^{18,26}. The probe line used to extract nvPM emissions sample was connected to a 3-way splitter using a 7.5 m long, 7.9 mm internal diameter thin-walled stainless steel tubing maintained 160°C. The nvPM sample was diluted with particle-free nitrogen gas via a Dekati ejector diluter and carried to the measurement suite along a 25 m long, 7.9 mm internal diameter, carbon-loaded and electrically grounded PTFE tube maintained at 60°C in accordance with SAE AIR6241. The nvPM number-based emissions were measured using an AVL Advanced Particle Counter, while nvPM mass-based emissions measurements were obtained using an Artium Laser Induced Incandescence and an AVL Micro Soot Sensor (MSS). Only the nvPM mass data obtained using the MSS is used in this analysis. The

particle size distributions of the nvPM, which are not specified in AIR6241, were measured using the Cambustion DMS500. The CO₂ concentration in the diluted nvPM line was measured using a LiCor NDIR detector. The nvPM emissions data are reported at standard temperature and pressure (273.15 K and 101.325 kPa), which is equivalent to mass concentration data reported via FOA3. All nvPM emission concentration data was corrected for dilution and thermophoretic loss in the sampling system. Measurement uncertainties in nvPM emissions were calculated using 1σ standard deviation of the average data.

Properties of test fuels

The two kerosene fuels used in this study were Jet A-1 and UCO-HEFA. The Jet A-1 was straight-run kerosene obtained from Air BP (Kingsbury, UK), while the UCO-HEFA was provided by SkyNRG (Amsterdam, NL). A GC x GC chemical analysis was used to quantify the paraffinic and aromatic chemical composition of the two fuels, a summary of which is shown in Figure 1. The figure shows the significant difference in the composition of the two fuels: the Jet A-1 contains a substantial fraction of cyclo-paraffins and aromatics, whereas these are much reduced for the UCO-HEFA fuel that is dominated by iso-paraffins.

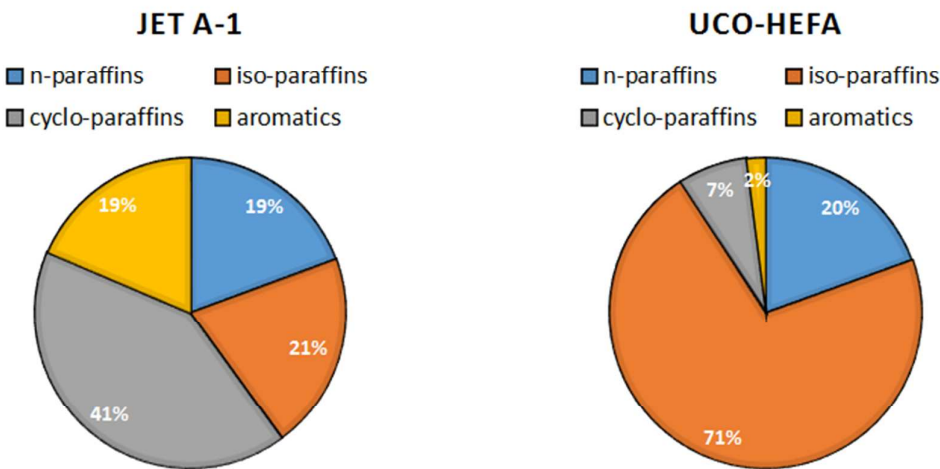


Figure 1. Summary of the GC x GC compositional analysis for the Jet A-1 and UCO-HEFA kerosene fuels that were used to formulate the test blends.

A number of Jet A-1 / UCO-HEFA kerosene fuel blends were formulated in-house through careful weighing and thorough mixing (blend ratios of 5%, 10%, 15%, 20%, 25%, 30%, 40%, 50%, 60%, 70%, 75%, 80%, 85%, 90%, and 95% by mass). The chemical composition of the fuels varied linearly with fuel blend ratio, and test fuel H:C ratio varied from 1.89 to 2.14 whilst aromatic content correspondingly varied from 19.2% to 1.8% by mass. The Jet A-1 and UCO-HEFA fuels were fully miscible and the blended fuels were formulated at least 48 hours prior to use. It is recognized that several of these blends are outside of current ASTM certification limits for HEFA fuel blends in operational aircraft, however these limits are no longer applicable to the now ground based APU used within this study. Further details of the fuel properties for neat Jet A-1 and UCO-HEFA fuels are given in Lobo et al.⁵².

By introducing the hypothetical concept of an aromatic - H:C ratio space, these fuels can be compared with fuels in the EDB, a world survey of the available JP8 fuels, and the nominal bounds for JP8 jet fuel. The specification for JP8, a military grade kerosene made to more exacting specifications than commercial jet fuel, is used in this context as a proxy, since H:C ratio is not defined for checklist Jet A-1. This comparison is shown in Figure 2.

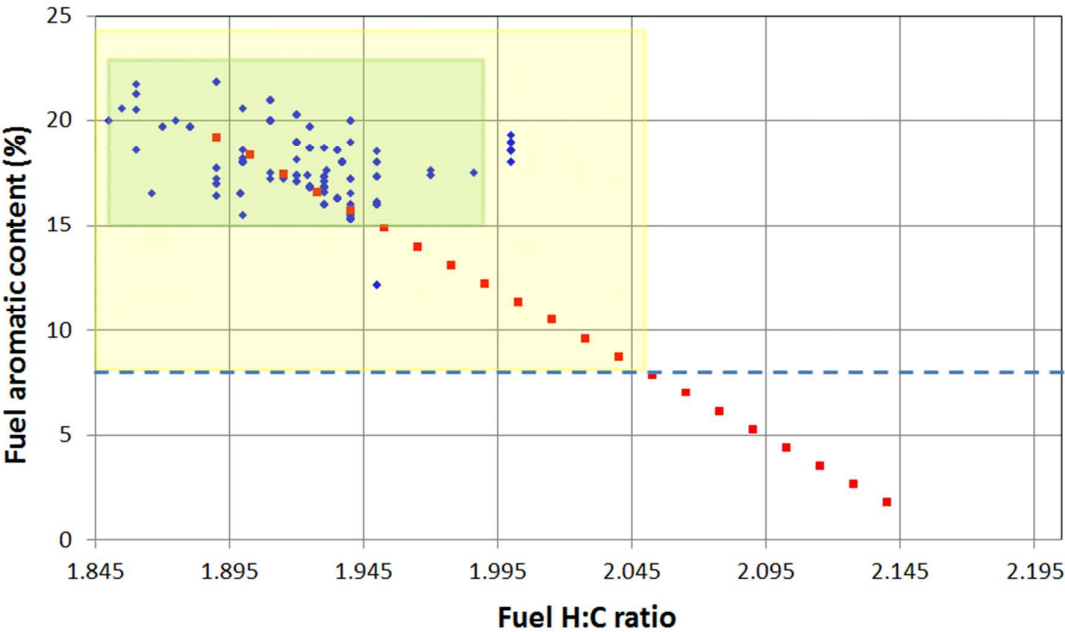
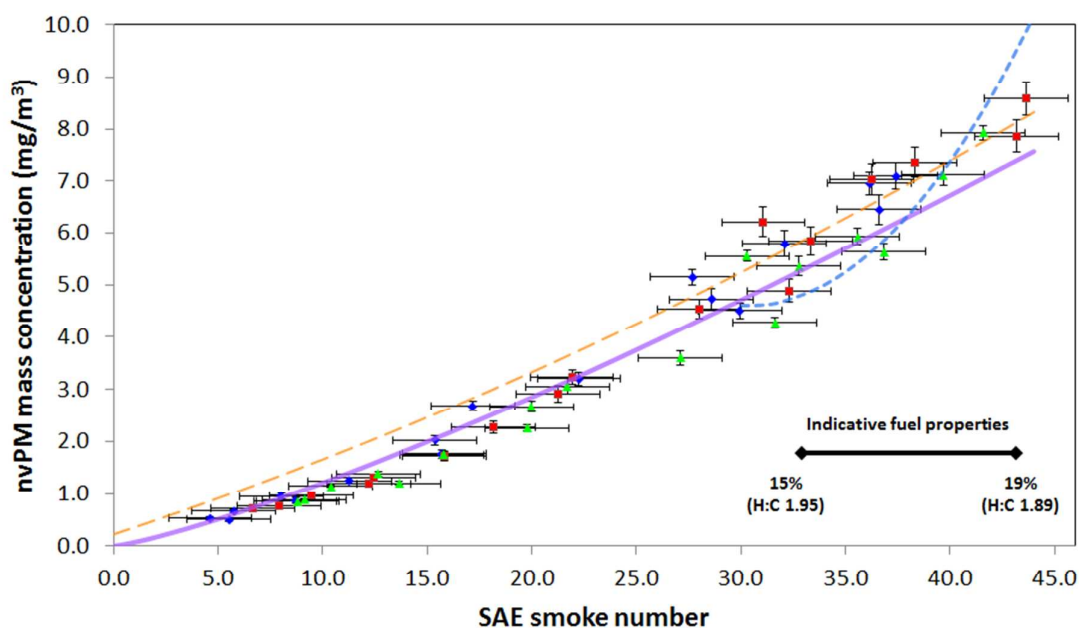


Figure 2. Comparison of aromatic content and H:C ratio of different fuels: Blue points: EDB engine certification data; Red points: Experimental fuel blends; Green shaded area: Bounds of ICAO engine test fuel specification¹⁷; Yellow shaded area: Bounds of PQIS world JP8 2013 survey⁴⁴; Chart area: Nominal bounds for JP8 jet fuel; Blue dashed line: ASTM D7566 minimum aromatic limit⁵⁴.

RESULTS AND DISCUSSION

Correlation between nvPM mass concentration and SN

Figure 3 shows the measured nvPM mass concentration, corrected for dilution and thermophoretic loss¹⁸, as a function of SN. The different colored data points in the plot indicate the three different engine conditions, and the BC mass concentration as a function SN predicted by FOA3 for both SN<30 and SN>30 are also overlaid.



342

343 Figure 3. nvPM mass concentration as a function of smoke number. Colored data points
 344 indicate the three different engine operating conditions: Blue=MES; Red=ECS; Green=NL.
 345 The C(BC) as predicted by FOA3 using SN data is overlaid: Purple line = FOA Equation 1
 346 (nominally applicable for SN<30); Dashed blue line = FOA Equation 2 (nominally applicable
 347 for SN>30); Dashed orange line = upper bound for Equation 1 generated using +3 SN error
 348 [Wayson et al.¹²]. An indication of the change in fuel aromatic content (H:C ratio) for ECS
 349 operating condition is inset.

350

351 The experimental data for the correlation between SN and nvPM mass concentration show
 352 close agreement with FOA3, particularly at SN<30. Furthermore this agreement is
 353 maintained over a wide range of kerosene compositions, and is largely independent of the
 354 engine operating condition. Lines of regression for the datasets representing the three engine
 355 conditions are practically coincident (not shown in the figure). The location of specific
 356 emissions data on the FOA3 curve is merely dependent upon the chemical properties of the
 357 fuel. Data points towards the left in Figure 3 represent measurements from fuel blends with

lower aromatic content and correspondingly greater H:C ratio. It is clear that the chemical composition of the kerosene has a significant impact on the tendency to form nvPM. Fuel aromatics have been identified as compounds that primarily influence the tendency to form soot aerosol during combustion^{45,46}, although it has long been suggested that fuel hydrogen content may be a more fundamental parameter that is independent of molecular structure^{58,59}. The data presented here cannot be used to differentiate between the impact of aromatics and the impact of H:C ratio since both vary linearly in the two component fuel blends. Experimental data using multi-component blends or surrogate fuels to adjust these parameters independently is necessary to explore their relative authority. The magnitude of the reductions in SN and/or nvPM emissions are comparable with data reported elsewhere for other gas turbine engines burning paraffinic fuels^{38,44,45,47,48}.

The nvPM mass concentration (C(nvPM)) and BC mass concentration (C(BC)) as defined by their respective measurement methodologies are not identical, and generally $C(\text{nvPM}) \geq C(\text{BC})$ since the former encompasses line loss correction factors that are not inherent in latter. The two standards are however closely related and these data support the supposition that C(nvPM) can be estimated from FOA3, but more significantly, that FOA3 can be used with alternative fuel blends of varying chemical composition without adaption or modification.

The data indicate that the relation between C(nvPM) and SN is foremost represented by FOA3 equation (1), even at $\text{SN} > 30$. Using all data points in Figure 3 and a power law fit to be consistent with FOA3 equation (1), the line of regression is given by:

$$C(\text{nvPM}) = 0.048 (\text{SN})^{1.35} \quad (3)$$

Whilst using a constrained range of data points up to $\text{SN} < 30$, the line of regression is given by:

$$C(\text{nvPM}) = 0.058 (\text{SN})^{1.27} \quad (4)$$

with the correlation coefficients of $R^2 = 0.979$ ($n=51$) and $R^2 = 0.965$ ($n=33$), respectively (to simplify the representation of data, these lines of regression are not included in Figure 3).

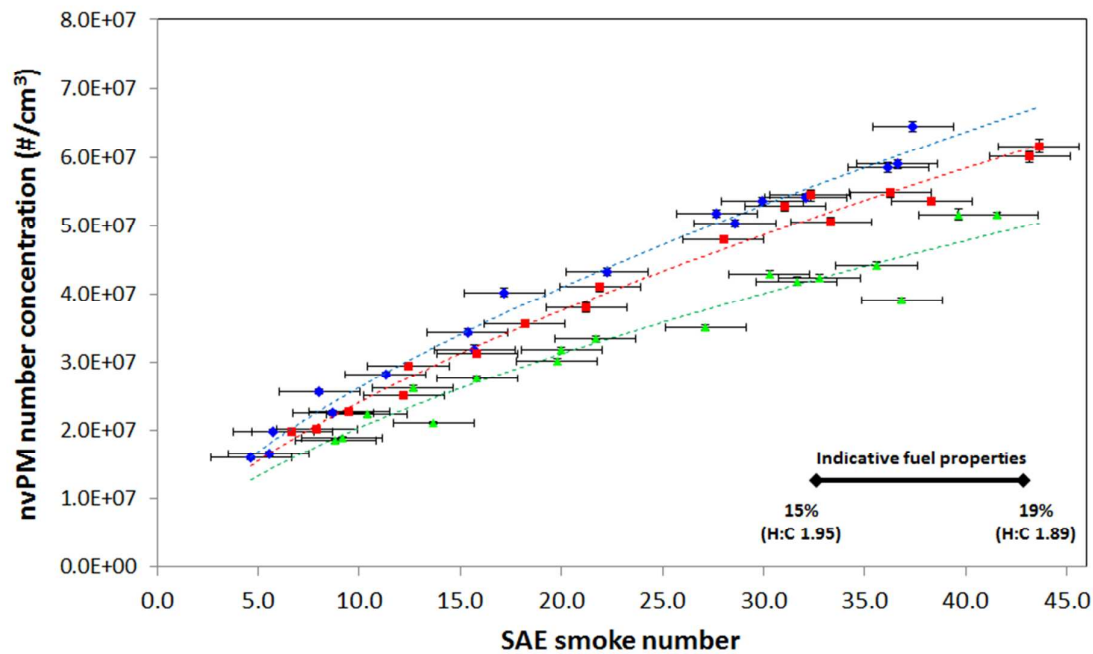
Figure 3 also shows a marker to indicate the 15% - 19% fuel aromatic range for the ECS engine operating condition (markers for other engine conditions are of comparable magnitude but offset relative to the SN axis). This marker corresponds to the mid-range and the lower bound for aromatic content in the ICAO specification for fuel to be used for aircraft engine certification testing. For this modest shift in fuel composition, the SN decreased by 30% and C(nvPM) decreased by 45%. Hence the fuel-induced uncertainty in EDB SN or C(nvPM) derived through FOA3 is potentially twice this number, when considered in respect of the limits of fuel used for engine certification testing¹⁷ and typical commercial fuel variability⁴³. SN data for a particular engine in the EDB is strictly only correct for the stated certification test fuel and will increase or decrease in magnitude for fuel of different chemical composition.

The data suggest that for engines with relatively large reported SNs the fuel-induced uncertainty could be significant and markedly greater than the nominal ± 3 SN uncertainty associated with the measurement of SN, whilst for engines with relatively small reported SNs the fuel-induced uncertainty will be captured within this same ± 3 SN measurement uncertainty. The proportional reduction in nvPM mass are consistent with data reported by Brem et al.⁵⁵ in a study evaluating the impact of fuel aromatic content on nvPM emissions from an in-production gas turbine engine.

nvPM number concentration and size distribution

Figure 4 shows the measured nvPM number concentration corrected for dilution and thermophoretic loss¹⁸ as a function of the measured SN. Measurement uncertainties are as

406 previously described, and similarly the different colored data points in the plot indicate the
407 three different engine conditions.



408
409 Figure 4. nvPM number concentration as a function of smoke number. Colored data points
410 indicate the three different engine operating conditions: Blue=MES; Red=ECS; Green=NL.
411 An indication of the change in fuel aromatic content (H:C ratio) for ECS operating condition
412 is inset.

413
414 Data points towards the left in Figure 4 represent measurements from kerosene fuel blends
415 of lower aromatic content and show a progressive reduction in the nvPM number
416 concentration. In this case there is some distinction between lines of regression for the three
417 datasets (shown in the figure) indicating that the relation between nvPM number
418 concentration and SN may be dependent upon the engine operating condition.

419 Figure 4 also shows a marker to indicate the 15% – 19% fuel aromatic range for the ECS
420 engine operating condition corresponding to the mid-range and the lower bound for aromatic
421 content in the ICAO specification for fuel to be used in aircraft engine certification testing.

For this shift in fuel composition, SN decreased by 30% and the nvPM number concentration decreased by 22%. This would suggest that nvPM number concentration is also a strong function of fuel composition, an observation that is consistent with data reported elsewhere^{52,55}.

The nvPM size distribution parameters of geometric mean diameter (GMD) and geometric standard deviation (GSD) for the fuel blends tested at each of the three APU operating conditions are shown in Figure 5. The nvPM exhibited a characteristic lognormal size distribution, which narrows and shifts the geometric mean diameter to smaller sizes as the aromatic content of the fuel blend is decreased (correspondingly increased H:C ratio). For a given fuel, the succession of nvPM GMD tracked the sequence NL>ECS>MES. Overall the GMD varied from a minimum of 22nm for 1.8% aromatic fuel in the MES engine condition to 42nm for 19.2% aromatic fuel in the NL engine condition. The corresponding GSD ranged from 1.58 to 1.79. Hence on the microscopic scale, the fuel-induced reduction in the mass of emitted nvPM corresponds to the emission of fewer and smaller units of particulate matter. These data are consistent with those reported for other gas turbine engines burning conventional and alternative fuels^{21,26,47,49,50}.

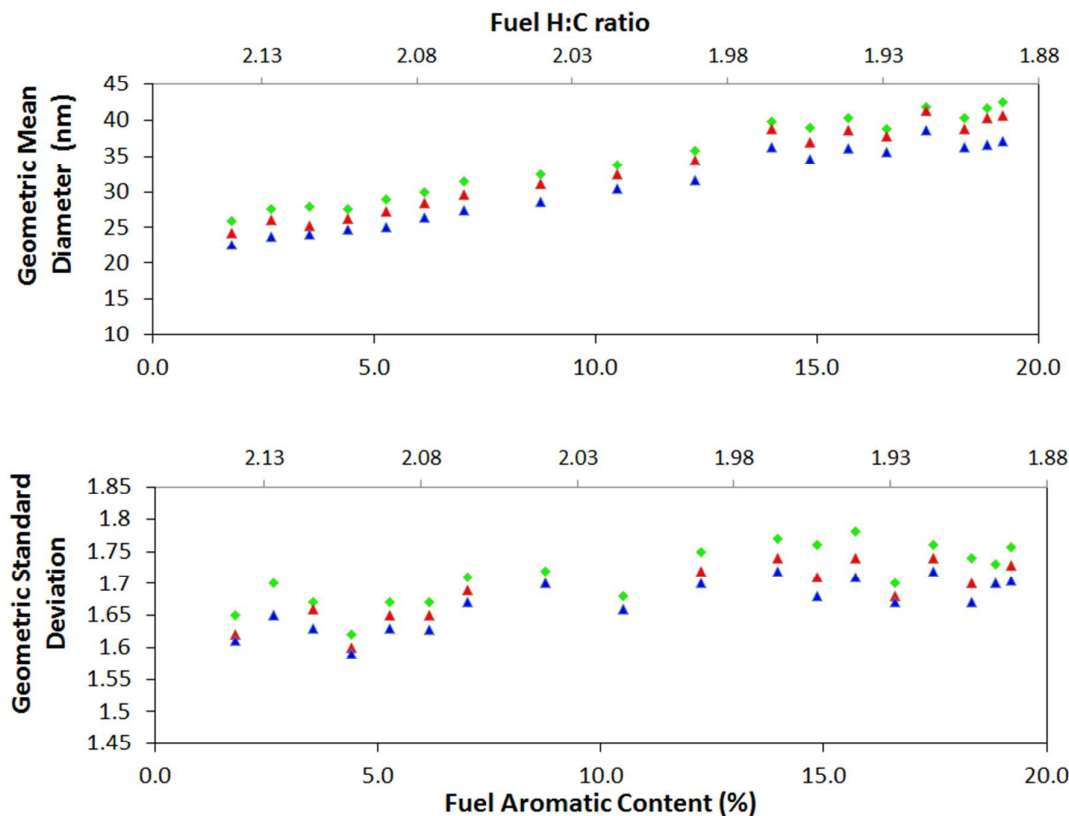


Figure 5. Geometric mean diameter and geometric standard deviation for nvPM emission from selected fuel blends. Colored data points indicate the three different engine operating conditions: Blue=MES; Red=ECS; Green=NL. In both cases the upper secondary axis shows the corresponding fuel H:C ratio.

When these nvPM GMD data are considered in relation to the C(nvPM) in Figure 3, it is evident that the small nvPM with GMD ~ 22nm that are characteristic of modern aircraft engine emissions, do not result in significant deviations in the FOA3 estimation of mass concentration. Previously Stettler et al.¹⁶ published data that appears to show that the relation between SN and C(nvPM) deviates significantly from FOA3 for nvPM with a GMD of the order of 20nm, with deviations of up to a factor 3. The data presented here do not support this finding. The data presented by Stettler et al.¹⁶ do not represent a fair comparison with the FOA3 method in three principal ways. Firstly, the applied methodology did not have a direct

measurement of nvPM mass concentration, but rather estimated it from size distribution and effective density measurements. In the current work, we directly measure nvPM mass. Secondly, Stettler's experiments were based on laboratory measurements of propane diffusion flame combustion, and the black carbon generated from a propane burner is not a model particle that is representative of the soot aerosol produced by a gas turbine engine. Propane burners produce black carbon via a different mechanistic route (as chemically dissimilar) that result in high EC fraction particulate matter with different physical and chemical properties. This assertion is supported by experimental data from Durdina et al.⁵⁶. Thirdly, the SN measurement methodology employed by Stettler was not comparable with the methodology that has been used to populate the data in the ICAO EDB. The use of a catalytic stripper to remove the semi-volatile OC from the line is not compliant with SAE ARP1179d⁶ and will result in a relatively 'clean' source of soot aerosol to be impingent upon the SN filter. The impact of volatiles to the measurement of SN was demonstrated by Rye et al⁶⁰.

The data in Stettler et al.¹⁶ do demonstrate that a 'clean' black carbon from a propane burner is captured with a progressively decreasing efficiency as the geometric mean diameter is reduced. However, the extrapolation that these data are applicable to the emission of nvPM from an aircraft gas turbine engine cannot be justified because of the differences in both the modeled source for BC / nvPM and the measurement methodologies employed. This is important since Stettler et al.¹⁶ claim that the FOA3 significantly underestimates aircraft emissions of BC by a factor of 2.5 – 3 for SN ≤ 15 , and consequentially, propose a factor ~ 3 upwards revision of aircraft BC radiative forcing which would make it equivalent to $\sim 1/3$ of the aviation CO₂ radiative forcing⁵⁷. On the basis of the measurements presented here and critique of the Stettler et al¹⁶ methodology, such conclusions and extrapolations cannot be supported.

Significantly, this work develops a comparative framework between current and future regulatory standards for the measurement of soot aerosol from a gas turbine that incorporates the quantitatively distinct emission from the combustion of alternative fuels, and places these within the ICAO endorsed and widely accepted FOA3 methodology. With typical engine lifetimes exceeding 20 years, older legacy engines will continue to contribute to overall emission levels and so both SN as a surrogate measurement of BC, and FOA3 as a vital assessment tool, may not be fully transitioned for some years to come. The importance of fuel composition and the impact of its attendant variability may be particularly acute in the application of EDB data to air quality modeling and the development of emission inventories.

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ABBREVIATIONS

BC, Black Carbon; C(BC), Concentration of Black Carbon; C(nvPM), Concentration of non-volatile Particulate Matter; EDB, Emissions Data Bank; FOA3, First Order Approximation (version 3); GMD, Geometric Mean Diameter; GSD, Geometric Standard Deviation; ICAO, International Civil Aviation Organization; LTO, Landing and Take Off; nvPM, non-volatile Particulate Matter; SN, Smoke Number; UCO-HEFA, Used Cooking Oil derived Hydrotreated Esters and Fatty Acids.

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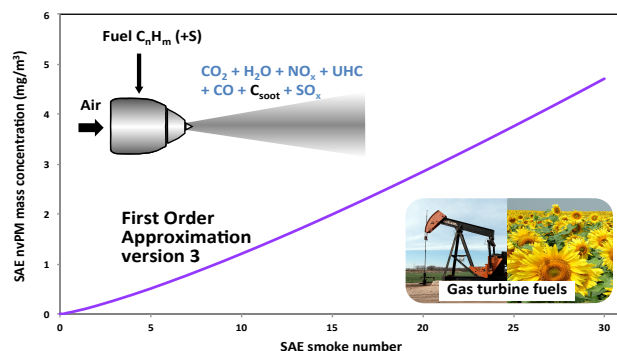
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